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Survey on Photon Activation Analysis Using the NRL Linac

W. L. BENDEL AND S. K. NUMRICH

Linac Branch
Nuclear Physics Division

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NAVAL RESEARCH LABORATORY
Washington, D.C.

CONTENTS

ABSTRACT

Experiments show that microgram to nanogram quantities of carbon, nitrogen, oxygen, or fluorine can be detected using photon-activation analysis. Irradiation with Linac bremsstrahlung is followed by coincidence counting of annihilation radiation. The minimum detectable amount of an element is strongly dependent on the material involved and the irradiation energy. Similar amounts of some other elements can be detected.

PROBLEM STATUS

This is a feasibility report on a method of analysis. Further work will depend upon funding for specific applications.

AUTHORIZATION

NRL Problem H01-09
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INTRODUCTION

Among the elements most difficult to detect in trace amounts are the light elements which are present almost everywhere. Air, water, and all organic material are made up primarily of four elements — hydrogen, carbon, nitrogen, and oxygen. To detect any of these elements as impurities, unusual care must be taken not to expose the sample to contamination during the analysis.

Activation analysis is a useful tool for detecting trace elements. After activation, the sample may be freely exposed to contaminating materials. For the analysis of surfaces or films, short-range bombarding particles, such as ions from a Van de Graaff accelerator, are indicated. For essentially uniform activation of materials, more penetrating radiation must be used, such as bremsstrahlung (gamma radiation produced from high-energy electrons), neutrons, or perhaps electrons. If desired, the surface may be removed after activation and before counting. The nuclear reaction to be employed is primarily determined by the nuclear properties of the element to be measured and of the other expected constituents of the sample.

Under proper conditions, one may irradiate a sample and then measure the element of interest from the induced radioactivity. Under other conditions, it will be necessary to treat the sample chemically after irradiation to separate the desired radioactivity from unwanted radioactivities. It should be noted that a particularly insidious reaction — often a minor reaction on a major constituent — is that which produces the same radioactive isotope as the desired activity. This case cannot be solved by chemical separation after irradiation, but may often be managed by a judicious choice of irradiation conditions, such as the bombarding energy.

The simplest detection system merely records radioactivity vs time. Better systems are more selective and respond only to certain types of radioactivity, sometimes being essentially specific to one element. As the greatest sensitivity usually goes with the minimum selectivity, one must tailor the system to the problem at hand.

PHOTONEUTRON REACTIONS AND POSITRONS

Three of the four light elements mentioned above — all but H — produce abundant radioactivity by the (gamma, neutron) reaction. This is the most probable reaction produced by the bremsstrahlung from an electron accelerator, although the (gamma, proton) reaction is comparable for light nuclei.

As shown in Table 1, about 99% of the atoms of C, N, or O are of the lightest stable isotope. Bombardment with bremsstrahlung produces positron activities of relatively convenient lifetime by the (γ, n) reaction on these isotopes. Fluorine, with similar properties, is also listed. The (γ, p) reactions on these four elements produce stable isotopes from the abundant isotopes and isotopes with undesirable half-lives (either 5730 years or less than 8 sec) from the rare isotopes. The (n, γ) reactions — produced by a reactor or as a secondary reaction here — also produce only radioactivities with inconvenient lifetimes, and these only from the rare isotopes and with small cross sections.

Table 1
Radioactivity Following the (γ,n) Reaction on the Isotopes of Four Elements

Element	Parent Isotope	Abundance (%)	(γ,n) Threshold	Daughter Isotope	Half-life	Radiation
Carbon	^{12}C	98.89	18.735 MeV	^{11}C	20.4 min	β^+, no γ
	^{13}C	1.11	—	^{12}C	Stable	
	^{14}C	$\lesssim 10^{-10}$	—	^{13}C	Stable	
Nitrogen	^{14}N	99.63	10.557 MeV	^{13}N	10.0 min	β^+, no γ
	^{15}N	0.37	—	^{14}N	Stable	
Oxygen	^{16}O	99.76	15.676 MeV	^{15}O	124. sec	β^+, no γ
	^{17}O	0.037	—	^{16}O	Stable	
	^{18}O	0.204	—	^{17}O	Stable	
Fluorine	^{19}F	100	10.433 MeV	^{18}F	110. min	β^+, no γ

It will be observed that each of these elements produces radioactivity without gamma rays. The detection of these activities is thus a matter of detecting positrons. The general approach, and the specific details of the present work, are given in Appendix A.

Many other elements may be similarly detected. Appendix B lists the positron activities produced by the (γ,n) reaction on stable isotopes. Other elements may be detected by nonpositron radioactivities following the (γ,n) or (γ,p) reaction or by radioactivities following reactions due to Linac-produced neutrons.

MINIMUM DETECTABLE MASS

Apparatus was set up in November 1969 to measure positron radioactivity produced by the NRL Linac. The counting system is described in Appendix A. The irradiation system has been reported by Wilkniss and others* and is briefly described in Appendix C. After preliminary tests and runs, specific conditions were adopted and a series of runs was made in December.

The results are given in Appendix C. The activity produced is a sensitive function of the Linac energy and is expressed in terms of a mass M_2. This is the mass of the element required to produce a net count of two standard deviations under specified conditions.

The mass M_2 is plotted as a function of ΔE in Fig. 1, where ΔE is the difference between the Linac bombarding energy and the reaction threshold energy. The letters in this figure show the oxygen, nitrogen, fluorine and carbon results from Appendix C. The limited data suggest that M_2 is proportional to $(\Delta E)^{-3.8}$, and a line with this slope is shown through the nitrogen points.

*Wilkniss, P.E., Hoover, J.I., and Leighton, R.E., Nucl. Inst. and Methods 56:120 (1967).

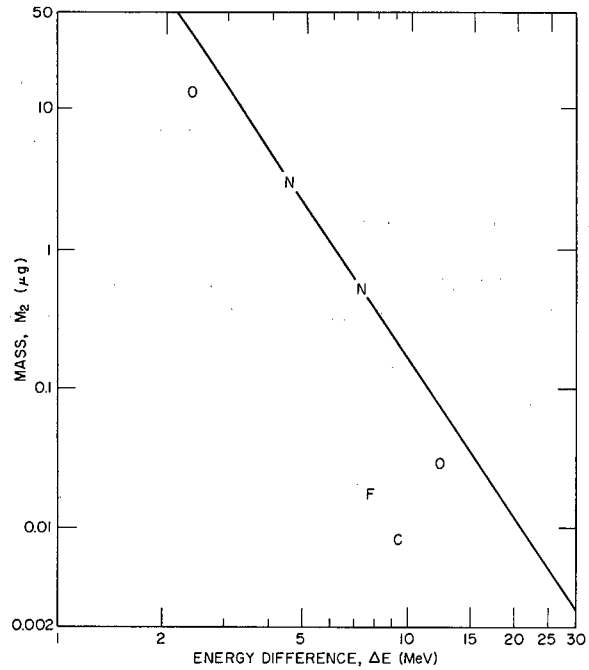

Fig. 1 - Minimum detectable amount M_2 of an element as a function of ΔE, the Linac energy minus the reaction threshold energy.

Improvements in apparatus and method, discussed in Appendix A, particularly in intercrystal shielding and the use of upper limits of pulse height, can decrease M_2 somewhat. Interfering activities and the need for chemical separation may dictate a minimum detectable mass much greater than the ideal M_2 in a given case.

These experiments show that submicrogram quantities of carbon, nitrogen, oxygen, or fluorine can be detected using photon-activation analysis. The method can be used for trace analysis for a limited number of other elements, as shown by the tables in Appendix B. Many other elements produce lesser positron activities and can be detected by this method only when present in much larger quantity.

Appendix A

DETECTION OF POSITRONS

METHOD OF DETECTION

While gross positron activities may be observed by many means, the measurement of trace elements requires the use of an especially sensitive and selective method.

In the cases of Table 1, there is no excited level of the daughter nucleus to which the radioactive nucleus is energetically capable of decaying; the decay is entirely to the ground state, by positron emission or by electron capture. As the electron capture process is very difficult to observe in light nuclei, the positrons must be detected. The most sensitive method is to observe the annihilation radiation rather than the short-range positrons themselves. A positron will quickly "combine" with a normal electron, annihilating both and usually producing two 511-keV photons traveling in opposite directions. This penetrating radiation, with its singular angular correlation, is the common indicator of positrons. However, aside from lifetime, it is the same radiation whether from C, N, O, F, or any other source of positrons.

The annihilation radiation is most readily detected by employing thallium-activated sodium iodide crystals. In order to discriminate against other photons of similar energy, two NaI crystals are used on opposite sides of the irradiated sample. The geometry of the apparatus used for the present work is shown in Fig. A1. A diagram of the apparatus and circuitry appears in Fig. A2.

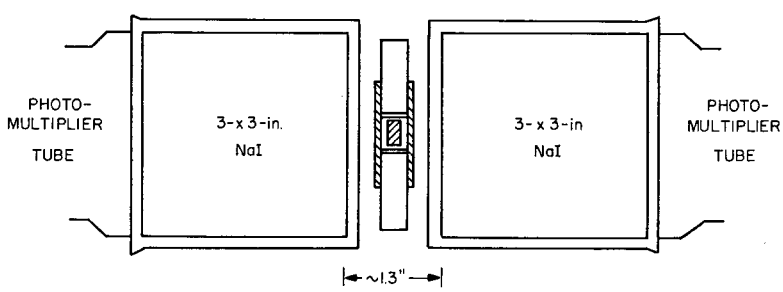

Fig. A1 - The sample is placed in a 0.375-in.-thick Bakelite holder with sides of 0.063-in. aluminum. A Bakelite lid, not shown, covers the sample. In the work reported here, the NaI housings were placed 1.0 in. apart, symmetrically about the sample holder.

Any of three things may happen if an annihilation photon enters a NaI crystal. It may be transmitted, it may be totally absorbed (in one or more steps), or it may be partially absorbed with a secondary photon escaping. A pulse will be produced with amplitude proportional to, and usually expressed as, the energy deposited in the crystal. The pulse height distribution will consist of a rather flat spectrum from 0 to about 330 keV,

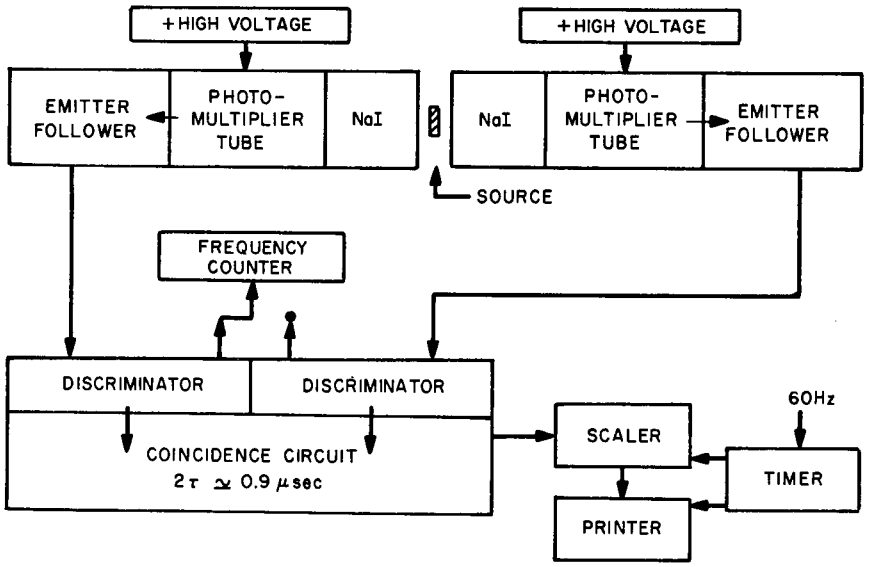

Fig. A2 - Apparatus and circuitry used in the present experiment. The number of coincidence counts is printed at the time interval chosen. The singles counting rate in either channel may be read with a frequency counter.

a peak at 511 keV (about 50 keV wide), and only a few pulses between 330 keV and the peak.

When one photon is initially headed for one NaI crystal, the other photon MUST be headed for the other NaI crystal if the NaI's are symmetrically placed about the site of the annihilation. In many cases, therefore, a pulse in one crystal will be coincident in time with a pulse in the other crystal. These coincidences are employed as the quantitative measurement of the positron activity present.

Other activities, whether in the sample or in the background, will also produce coincidences. In addition to radioactive decay involving two or more gamma rays in cascade, a decay with only one gamma ray can cause some coincidences due to backscattering from one crystal to the other. A strong activity will also produce accidental coincidences. These problems may be reduced by appropriate choice of geometry and discriminator level.

ACTIVATIONS

A series of activations was made with the NRL Linac. The irradiation conditions and results are given in Appendix C. The discriminators were set to respond to pulses corresponding to at least 200 keV. Coincidence counts were obtained as a function of time, and the amount of radioactivity of predetermined half-life was extracted.

VARIATIONS IN METHOD

The techniques and apparatus employed here can be altered in innumerable ways. Let us consider the effect on sensitivity of some of these alternatives.

Irradiation

It is clear from the data (Appendix C) that the activity is a sensitive function of energy. In an otherwise inert sample, the maximum attainable beam current at a bombarding energy of 40 to 50 MeV will produce considerably better sensitivity than obtained for the cases of Appendix C. A nanogram of material should be detectable in some cases, but the increase in unwanted activities will make more chemical compositions unsuitable.

The location of the sample during irradiation is good. A small sample could be somewhat nearer the tantalum bremsstrahlung radiator and in a more intense radiation field. A sample somewhat further from the tantalum would be more uniformly irradiated.

Transfer and Preparation

The apparatus could be relocated to cut some seconds from the 30-sec time interval between irradiation and counting. If the sample is irradiated in a capsule and then manually removed from the capsule, no radical shortening of time can be made. Entirely different techniques, with lesser sensitivity, can be used for lifetimes of a second or a microsecond. On the other hand, a longer interval is preferable if it permits useful chemical treatment. It is clear that chemical processing which is practical for a long-lived activity such as ^{18}F could be much too time-consuming to be useful for ^{15}O.

Detection

By counting singles, rather than large-angle coincidences, one may obtain more counts from any activity. However, the increase for positron emitters is quite modest here (a factor of about 2 with discriminators at 200 keV) compared to the increase for background (a factor of about 100) and for most radioactivities.

The optimum NaI configuration would not be appreciably better than our present apparatus. As 3 in. of NaI absorbs 92% of annihilation quanta, no significant improvement is attainable with larger crystals. The fraction of the pulses in the 511-keV peak also would show only slight improvement for larger crystals. Increased sensitivity to other radiation largely nullifies the slight effect on the desired radiation.

Improvements can be made by more stringent requirements on the pulse heights. By employing more complex apparatus with an upper limit of about 580 keV on each channel, one would eliminate many coincidences from background (particularly cosmic rays) and interfering radioactivities. The optimum lower limit is not so clearly determined. The value used here — 200 keV — together with the geometry, permits many coincidences due to one gamma ray, with Compton scattering from one NaI to the other. A bin centered at 511 keV in each channel would be relatively sensitive to positrons, but would have reduced absolute sensitivity. Calculations with the positron and background data show little net improvement for other lower limits under the conditions employed, i.e., with no upper limit.

If one employs a large germanium solid-state radiation detector with a multichannel analyzer, one can observe sharply resolved gamma-ray lines. Although the photopeak efficiency is markedly less than for NaI, germanium has a clear advantage in identifying activities with several distinctive gamma-ray energies. Its merit relative to NaI is probably lowest for the activities considered here — activities having no gamma rays other than annihilation radiation with its distinctive angular correlation and equal energies of the coincident quanta.

By simply inserting a 1- by 4- by 4-in. piece of lead between the crystals, the background coincidence rate is decreased by a factor of 4, primarily by preventing intercrystal scattering. While this shielding would also forbid real positron-produced coincidences, a lead shield with a hole of about 0.75-in. diameter is feasible and does almost as well. As in the present system, the sample should still be surrounded with sufficient light-element material to stop most positrons with little attenuation of the 511-keV quanta.

The solid angle to be employed is a compromise of many factors. Maximum counts and maximum selectivity in angle are incompatible. As background coincidences went down no faster with distance than annihilation coincidences, the crystals were used close together. With interfering activities, improved shielding, and/or different pulse height requirements, smaller solid angles may be indicated.

Appendix B

POSITRON ACTIVITIES PRODUCED BY (γ,n) REACTIONS ON STABLE ISOTOPES

The elements which produce positron (β^+) radioactivities by the (γ,n) reaction when irradiated with bremsstrahlung are listed in Tables B1 and B2. In employing an electron accelerator to produce bremsstrahlung of ~10 MeV to ~30 MeV, one produces many activities by other reactions. It is rare when any reaction other than (γ,n) yields a positron activity useful for the detection of impurities. Many reactions, however, can produce significant unwanted β^+ activities in the major constituents of a sample. These tables, then, are intended to show which elements can be observed in small quantities. They also show many weak β^+ activities, but do not include all possible interfering cases.

With bremsstrahlung irradiation, reactions such as (γ,p), (n,γ), and $(\gamma,2n)$ will occur. In those rare cases where (γ,p) produces a β^+ activity (Kr, Ru, and Ce), the (γ,n) reaction produces a more useful β^+ activity. In those rare cases where the secondary reaction (n,γ) – due to the many neutrons produced in and near the sample – yields a positron activity (Cu is the lightest), the same radioactivity is produced by (γ,n) on a relatively abundant isotope of the same element. The $(\gamma,2n)$ reaction will always have a high threshold and low yield, making it an unlikely means of detecting impurities.

The tables below include a figure of merit D for detectability using the method considered here. This quantity is

$$D = \frac{20\ \text{MeV}}{B}\ I\ F\ H,$$

where I is the isotopic abundance, B is the neutron binding energy, both of the parent isotope; F is the fractional decay of the product isotope by β^+; and H is a factor dependent upon half-life. No attempt has been made to incorporate the (γ,n) cross section into this equation, as it is a function of energy and is often not known.

The threshold energy for a (γ,n) reaction is slightly greater than the neutron binding energy B because momentum must be conserved in the reaction. The relationship is

$$\text{Threshold} = B\left(1+\frac{B}{2M}\right),$$

where M is the total energy (rest mass) of the initial nucleus.

The ideal half-life for the present apparatus is about an hour. Longer-lived activities require excessive Linac irradiation time as well as counting time, and encounter larger background drifts during counting. We thus want a factor λ, the decay constant, in H. Both the intensity and half-life of short-lived activities are difficult to measure, and a minimum transport time of about 30 sec is required from irradiation to counting. We therefore introduce a factor $e^{-\lambda T_0}$. The quantity obtained still does not fit the subjective impression of H as a function of λ. We finally adopt the relationship

Table B1
Elements for Which the β^+ Activity Following the (γ,n) Reaction is a Good Means of Detection; i.e., $D > 0.10$

Isotope	Abundance I (%)	B (MeV)	$T_{1/2}$	β^+ (%)	D	γ per Decay	γ Energy (MeV)	Main γ	Remarks
^{12}C	98.8	18.720	20.3 m	99.8	0.47	0			
^{14}N	99.6	10.553	10.0 m	100	0.63	0			
^{16}O	99.7	15.668	124 s	100	0.15	0			
^{19}F	100	10.430	110 m	97	0.80	0			
^{31}P	100	12.312	2.5 m	99+	0.22	0.005	2.16		
^{35}Cl	75.5	(12.635)	32 m*	99	0.28	0.77	0.64-4.10	2.14	G:1.56s:β^+
^{39}K	93.2	12.047	7.71 m	99+	0.22	0.02	1.65-2.79	2.79	0.95s*:β^+
^{45}Sc	100	11.319	3.92 h	94	0.21	1.01	1.16-2.67	1.16	2.44d*:EC
^{63}Cu	69.1	10.841	9.8 m	98	0.41	0.009	0.66-2.24	equal	
^{65}Cu	30.9	9.910	12 h	19	0.01	0.005	1.34	1.34	38% β^-
^{64}Zn	48.9	11.855	38.4 m	94	0.41	0.145	0.76-2.9	0.67	
^{69}Ga	60.2	10.324	68.3 m	88	0.51	0.058	0.8-2.32	1.08	
^{79}Br	50.5	10.699	6.5 m	92	0.23	0.14	0.61-1.31	0.613	
^{81}Br	49.5	10.161	17.6 m	3.6	0.005	0.085	0.62-1.33	0.618	4.4h*:100% IT
			4.4 h*	(3.6)	0.0038	2.1	0.04-1.33	0.04, 0.05	IT
^{107}Ag	51.35	9.531	24 m	60	0.31	0.35	0.51-2.71	0.512	8.4d*:EC
^{109}Ag	48.65	9.182	2.42 m	0.28	0.0004	0.026	0.43-1.01	0.63	97.8% β^-
^{121}Sb	57.25	9.250	16 m	44	0.11	0.013	1.17	1.17	5.8d*:EC
^{123}Sb	42.75	8.975	2.8 d	0.006	0.0001	0.868	0.564-1.26	0.564	4.2m*:IT
^{141}Pr	100	9.386	3.39 m	50	0.18	0.049	0.11-2.53	1.59	

Table B2
Elements Which Yield a β^+ Activity by the (γ, n) Reaction but Which are not Easily Detected by the Method Considered Here; i.e., $D < 0.10$

Isotope	Abundance I (%)	B (MeV)	$T_{1/2}$	β^+ (%)	D	γ per Decay	γ Energy (MeV)	Main γ	Remarks
^{20}Ne	90.92	16.865	17.5 s	100	0.004	0			
^{23}Na	100	12.418	2.60 y	90	$73/10^6$	1.0	1.275	1.275	
^{24}Mg	78.6	16.532	12.1 s	100	0.0008	0.09	0.439	0.439	
^{27}Al	100	13.057	6.4 s*	100	0.00001	0			G:7.4×10^5y:β^+,EC
^{28}Si	92.18	17.175	4.2 s	100	$25/10^8$	0			
^{32}S	95	15.092	2.7 s	100	$2/10^{10}$	0.011	1.27	1.27	
^{36}Ar	0.337	15.252	1.83 s	100	$<1/10^{10}$	0.07	1.18-1.73	1.19	
^{40}Ca	96.97	15.619	0.87 s	100	$<1/10^{10}$	0			
^{46}Ti	8.0	13.192	3.09 h	84	0.031	0.008	0.718-1.66	0.718	
^{50}Cr	4.31	12.930	41.9 m	94	0.033	1.02	0.063-0.15	0.089	
^{54}Fe	5.84	13.619	8.5 m	88	0.023	0.49	0.38- ?	0.38	
^{58}Ni	67.7	12.195	36 h	47	0.017	1.14	0.13-1.91	1.36	270d:EC
^{59}Co	100	10.467	71 d	15	0.0002	1	0.810-1.67	0.810	9h*:IT
^{70}Ge	20.55	11.529	38 h	35	0.004	3.63	0.237-2.02	1.107	
^{74}Se	0.87	(12.112)	42 m 7.1 h	63 65	0.005 0.001	0.19 1.0	0.088-0.58 0.065-0.359	0.251	G? G?
^{78}Kr	0.354	11.870	1.19 h	80	0.0024	2.46	0.024-0.87	0.131	→ 5.7h:EC,β^+;4.2m*:IT
^{80}Kr	11.56	11.507	34.9 h	8	0.00026	2.41	0.044-1.33	0.6, 0.398	55s*:IT
^{85}Rb	72.15	10.475	33 d	21	0.0004	0.766	0.88-1.9	0.88	
^{84}Sr	0.56	11.580	32.4 h	20	$7/10^5$	1.05	0.04-0.755	0.755	→ 83d:EC

(Table continues)

Table B2 (Continued)

Isotope	Abundance I (%)	B (MeV)	$T_{1/2}$	β^+ (%)	D	γ per Decay	γ Energy (MeV)	Main γ	Remarks
^{90}Zr	51.46	11.997	78.4 h	22	0.0014	1.01	0.91-1.71	0.91	4.2m*:94% IT
			4.2 m*	1.4	0.0018	0.97	1.51	1.51	
			IT	22	0.0013	2.01	0.59-1.71	0.59, 0.91	
^{92}Mo	15.86	12.580	15.5 m	94	0.048	0			→ G > 10^4y:EC
			66 s*	38	0.0026	0.88	1.31-1.64	1.64	43% → 62d*:EC
			IT	94	0.027	1	0.658	0.658	57% IT
^{93}Nb	100	8.844	10.61 d*	0.06	$6/10^6$	1.05	0.90-1.8	0.934	G > 350y
^{96}Ru	5.46	10.124	1.7 h	14	0.0065	1.98	0.34-1.4	0.34	→ 20h:EC
^{102}Pd	0.96	10.360	8.4 h	2.5	$6/10^5$	4.51	0.02-1.67	0.296	→ 4.5d*:EC,IT
^{106}Cd	55	10.870	55 m	1+	$1/10^5$	?	0.025-2.32	?	→ 40d:EC
^{108}Cd	0.88	10.334	6.5 h	0.28	$8/10^6$	1.03	0.03-1.2	0.093	→ 44.3s*:IT
^{113}In	4.23	9.427	14.4 m	22	0.0032	0.037	0.62-1.48	0.617	44% β^-
			20.7 m*	22	0.00016	1.04		1.55IT	
^{112}Sn	0.95	11.080	35 m	27	0.0001	0.098	0.75-2.32	0.75	→ 2.81d:EC
^{120}Te	0.089	10.283	15.9 h	5	0.00001	1	0.645-1.76	0.645	4.7d*:EC, $\beta^+ < 0.6\%$
^{124}Xe	0.096	10.500	2 h	18	0.00013	?	0.09-1.1	?	→ 13.3h:EC
^{133}Cs	100	9.038	6.6 d	0.6	0.00096	1.09	0.36-1.9	0.669	2.2% β^-
^{136}Ce	0.193	9.990	17.2 h	<1	$<4/10^7$	?	0.06-0.90	0.265	→ 19.5h:EC, 99.4% IT
^{138}Ce	0.250	9.470	9.0 h	≤0.009	≤0.00001	1.06	0.010-0.93	0.010	34.4h*:IT, 0.6% EC
			34.4 h*	(≤0.009)	$<2/10^6$	2	0.01-1.16	0.01, 0.25	
^{142}Nd	27.13	9.809	2.5 h	3	0.0058	0.04	0.14-1.3	1.13	63s*:IT
^{144}Sm	3.16	10.460	8.9 m	50	0.0094	0			64s*:IT
^{153}Eu	52.23	8.544	9.3 h*	0.011	$2/10^5$	~1	0.12-1.6	0.12	77% β^-; 96m* → G:12y: 0.021% β^+
^{156}Dy	0.0524	9.890	10 h	2.14	$3/10^6$	>1	0.06-1.6	0.06, 0.23	→ 5.6d:EC
^{162}Er	0.136	9.200	3.1 h	1	$4/10^6$	~1	0.06-1.97	0.826	→ 6s*:IT → 2.5h:EC
^{164}Er	1.56	8.795	75 m	0.004	$7/10^8$	0.001	0.3-1.2	0.43	

$$H = \frac{(100\,\lambda)\, e^{-(1.2\lambda)}}{1 + (100\,\lambda)^{1.5}},$$

where λ is in units of min^{-1}.

When isomers occur, we assume the (γ,n) cross section to be equally divided between them. When the positrons follow a cascade of half-lives, we employ the sum of the half-lives in the equation.

In addition to those already mentioned, we employ these abbreviations and symbols in the following tables:

EC	Electron capture	s	second
*	Isomeric state	m	minute
G	Ground state	h	hour
IT	Isomeric transition	d	day
→	Yields daughter state or activity	y	year
$T_{1/2}$	Half-life		

The primary source of data for Tables 1, B1, and B2 is the compilation by Lederer and others (B1). The "Nuclear Data Sheets" (B2,B3) and the work of Lauritsen and Ajzenberg-Selove (B4) are also utilized, plus the original publications in a few cases. Neutron binding energies are obtained from the mass tables and related data of Mattauch and others (B5).

A number of similar tabulations have been published for activation analysis utilizing photon irradiation, but not specifically for positron activities. Baker (B6) lists detection limits for a number of elements using a 30-MeV electron beam and one NaI detector. Lutz (B7) calculates specific activities for 66 (γ,n) reactions, plus nine (γ,p) reactions and one (γ,γ') reaction.

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Appendix C

ACTIVATIONS AND RESULTS WITH C, N, O, AND F

METHOD OF ACTIVATION

Irradiations with the NRL 60-MeV Linac have been made using the pneumatic "rabbit" system. This system has been described by Wilkniss and others (see footnote, p. 2).

The Linac electron beam is bent 45 degrees by a magnet and focused by a quadrupole magnet, the system transmitting a beam 5% wide in energy. The electron beam terminates in a thick tantalum disk. The bremsstrahlung produced in the tantalum passes through the sample position 1.0 in. further along the beam axis.

The material to be irradiated is placed in a cylindrical capsule, normally of aluminum, with an interior space 0.500 in. long and 0.312 in. in diameter. The filled capsule is propelled by compressed air through plastic tubing to a position in front of the tantalum target, where the capsule is rotated during irradiation. At the end of irradiation, compressed air is applied from the other side to return the capsule to the experimenter.

With a beam diameter of about 1/8 in. and a sample of 1/4 in. diameter, a rather small displacement of the beam will produce a considerable change in the activation produced. When the bombarding energy is not far above threshold, the distribution of electrons within the 5% bandpass may also be important. It is therefore doubtful whether beam current is an adequate monitor. A known sample should be irradiated with the unknown to calibrate each run.

RESULTS OF ACTIVATION

After preliminary runs and tests, data on four elements were obtained in December, 1969, using beam 2 of the NRL electron Linac. In all cases the sample consisted of plastic cylinders approximately 0.44 in. long, either one cylinder 0.250-in. in diameter or one to three cylinders 0.125 in. in diameter.

As the electron beam striking the bremsstrahlung-producing target is 5% wide in energy (nominal value ±2.5%), some of the irradiations were made at Linac settings about 3% below a (γ,n) threshold. This is a compromise between maximum irradiation for other activities and essentially no activation of the high-threshold activity. Some electrons will be accelerated to slightly greater energy and will be unable to reach the target, striking the beam aperture. Even though these electrons are stopped, some of the bremsstrahlung which they produce will irradiate the sample and a quite small activation will occur.

The discriminator in each channel was set to trigger for pulses corresponding to 200 keV or more. The irradiation time and counting intervals were chosen to fit the desired activity. These experiments are summarized in the top part of Table C1. Each counting rate is expressed in terms of C_1, the net count in the first interval, but the number is obtained from the known half-life and all of the counting data. The fraction saturated is the ratio of the activity produced to that for an infinite irradiation at the same conditions.

Table C1
Activation Analysis of Light Elements Using Linac Bremsstrahlung.*

	Sample						
	Nylon	Acrylic	Nylon	Acrylic	Teflon	Acrylic	Acrylic
Linac energy (MeV)	15.2	15.2	18.1	18.1	18.2	28.0	28.0
Electron beam (μA)	10	10	14	10	35	11	4.5
Irradiation time	21:00	3:01	7:00	3:00	3:00	1:00	0:09.6
Transfer time	1:01	0:30	0:34	0:47	6:10:24	0:30	0:31
Total mass (mg)	444.6	429.5	444.6	429.5	602.9	429.5	113.1
Background/min	100	100	90	90	91		96
Counting interval	3 min	30 sec	1 min	30 sec	10 min	30 sec	30 sec
	Element						
	Nitrogen	Oxygen	Nitrogen	Oxygen	Oxygen	Fluorine	Oxygen	Oxygen	Carbon
Mass of element (mg)	55.0	137.3	55.0	62.9	137.3	458.1	137.3	36.1	67.8
Fraction saturated (%)	76.7	63.6	38.4	90.4	63.4	1.876	28.5	5.22	0.542
C_1	220,000	(-3)	203,000	~31,000	31,000	1,560,000	Jammed	75,000	17,400
C_1 per μg	4.00	0	3.69	~0.49	0.226	3.41	—	2.08	0.257
Electron beam (μA)	18		30	30	30	30		42	42
Irradiation time	25 min		25 min	310 sec	310 sec	240 min		310 sec	51 min
Fraction saturated (%)	82.3		82.3	82.3	82.3	78.0		82.3	82.3
C_1 per μg	8.0		17.0	~0.97	0.97	1,260		308	364
Count time	25 min		25 min	310 sec	310 sec	240 min		310 sec	51 min
Counts per μg	35		209	~2.8	5.2	16,000		1,640	17,800
M_2 = element mass for 2σ net counts (μg)	2.9		0.5	~17	9	0.019		0.028	0.008

*The upper part of the table shows experimental conditions and results. The part below the dashed line shows the calculated sensitivity for an idealized case, using the conditions shown.

At 28 MeV, a beam of 42 μA was available using the maximum Linac pulse rate of 360 per second. Overactivation being anticipated, the first irradiation was performed with 180 pulses per second for one min. This proved to still be much too much. The second irradiation employed a smaller sample, a shorter irradiation, and 60 pps.

SENSITIVITY

The ultimate sensitivity of the system at the given energy is derived in the bottom part of Table C1. We employ the system in a near-optimum case, using the maximum Linac beam consistent with our information. It should be noted that the irradiations were short and the time spent on Linac tuning was usually not sufficient to properly maximize the beam.

In determining the sensitivity, we assume irradiation and counting times (except for F) of 2.5 half-lives each. A longer irradiation time would yield better results here, but not in a case with a longer-lived contaminant. The matter of optimum counting time is discussed in Appendix D. It is found that 2.5 half-lives yields near-optimum results for measuring intensity in almost all cases. However, a shorter counting time, about 1.5 to 2 half-lives, would be somewhat better for the small activities to be considered. The times assumed for fluorine are shorter than 2.5 half-lives because of the relatively large time involved.

We also assume that the counting starts 30 sec after the end of irradiation, that the background is 96 counts per minute, and that no other activities are produced. Thus, the bottom part of the table does not always apply to the material from which the data were obtained.

The last line in the table shows the mass M_2 necessary to produce a net count of two standard deviations, assuming background to be well determined. For an activity considerably less than background (as here), the required mass is increased by the factor $(1 + T/S)^{1/2}$ where T/S is the ratio of sample counting time to background counting time; see Appendix D.

It is seen that the minimum detectable mass of each of these elements is in the microgram range and decreases sharply as the bombarding energy is increased above the threshold energy. Let us consider an empirical equation of the form

$$M_2 = k\,(\Delta E)^{-n},$$

where k is a constant for a given element and ΔE is the difference between the Linac bombarding energy and the reaction threshold energy. The data here yield $n \cong 3.8$. The results of Table C1 are plotted in Fig. 1 in a manner exhibiting this relationship, with an $n = 3.8$ line through the nitrogen points.

The oxygen data from the 18.1-MeV irradiation of nylon are not well determined because they are obscured by the more intense and longer-lived nitrogen activity. Perhaps the discrepancy with the acrylic data at the same energy could be halved at best, but the value needed ($C_1 = 57{,}000$) to match the acrylic data is unreasonable. This discrepancy is an uncomfortably emphatic confirmation of the above-mentioned need for a calibration sample with each unknown.

Appendix D

OPTIMUM COUNTING TIME

Given an initial activity and other boundary conditions, what is the optimum counting time for determining the amount of that activity? The answer depends on many factors. How well is the background known? How intense is the activity relative to the background? What is the lifetime? What other radioactivities will influence the counting system?

We shall consider quantitatively the case with no interfering activities in the sample. This is not significantly different from the case with a very long-lived (relative to the activity of interest) interfering activity, except in the means of determining "background." It is also similar to the case with a very short-lived interfering activity, as this can be resolved with a rather short delay before counting. The simple case calculated here is different from the case of several activities with similar half-lives and from the case where the half-life cannot be assumed.

Let the counting rate be

$$\text{Rate} = A\, e^{-kt} + B\,, \tag{D1}$$

where A is the initial activity, k is the decay constant, t is the time, and B is the background. Upon integrating over a counting interval from 0 to T, one obtains

$$\text{Counts} = (A/k)\,(1 - e^{-kt}) + BT\,, \tag{D2a}$$

and

$$\text{Net counts} = \text{Counts} - BT = (A/k)\,(1 - e^{-kT})\,, \tag{D2b}$$

with a counting uncertainty $\Delta C = (\text{Counts})^{1/2}$.

The background B is determined from a separate set (or sets) of data, obtaining BS counts in time S. For unit time, one has

$$\text{Background} = \frac{BS \pm (BS)^{1/2}}{S} = B \pm (B/S)^{1/2} \tag{D3}$$

or

$$\Delta B = (B/S)^{1/2}.$$

The uncertainty in the net count is

$$\text{Uncertainty} = [(\Delta C)^2 + (T\Delta B)^2]^{1/2} = (\text{Counts} + BT^2/S)^{1/2}\,. \tag{D4}$$

Let X be the ratio of the uncertainty to the net count, and let the initial activity be n times the background, or

$$A = nB\,. \tag{D5}$$

We now have

$$X^2 = \frac{(nB/k)(1-e^{-kT}) + BT + BT^2/S}{(nB/k)^2 (1-e^{-kT})^2} . \tag{D6a}$$

This may also be written as

$$\frac{nB}{k}(1-e^{-kT})\, X^2 = 1 + \frac{kT(1+T/S)}{n(1-e^{-kT})} . \tag{D6b}$$

Now let us find the minimum value of X (for given A, B, k, and S) by taking the derivative with respect to counting time T and setting that equal to zero. The solution is

$$n_0 e^{-kT} = 1 + \frac{2T}{S} - \frac{2kT(1+T/S)}{e^{kT} - 1} . \tag{D7}$$

Note that it is impractical to solve for T, so we solve for n_0, the value of n for which a given T is optimum. For large activities (n large) and well-known backgrounds (S large), long counting times are best. For little activity and a poorly determined background, short counting times are indicated if one knows the half-life.

In practice, one must use a different approach. What value of kT is best in the general case, or in the range expected? With an initial activity a few times background and reasonable values of S, a counting time of 2.5 half-lives is a good choice. For $n \rightarrow \infty$ in Eq. (D6b), the ratio of $X_{2.5}$ to X_{min} (for a very long counting time) is 1.102. For $n \rightarrow 0$, $X_{2.5}/X_{min}$ is dependent on S and is 1.102 for S = 4.314 half-lives. It is thus seen that a choice of T = 2.5 half-lives is about as good (within 10%) as can be done for any case, provided sufficient background data are obtained.

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13. ABSTRACT

Experiments show that microgram to nanogram quantities of carbon, nitrogen, oxygen, or fluorine can be detected using photon-activation analysis. Irradiation with Linac bremsstrahlung is followed by coincidence counting of annihilation radiation. The minimum detectable amount of an element is strongly dependent on the material involved and the irradiation energy. Similar amounts of some other elements can be detected.

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